

Commissioning of a high-resolution collinear laser spectroscopy apparatus using a laser ablation ion source*

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Collinear laser spectroscopy is known as one of the powerful tools for the study of nuclear spins, electromagnetic moments and charge radii of the exotic nuclei. Aiming at studying these nuclear properties of unstable nuclei at the Beijing Radioactive Ion-beam Facility (BRIF) and the future High Intensity Heavy-ion Accelerator Facility (HIAF), we have firstly developed a collinear laser spectroscopy apparatus integrated with an offline laser ablation ion source and a laser system. The overall performances of this state-of-the-art technique and device have been commissioned by using the bunched stable ion beam. High-resolution optical spectra of $^{40,42,44,48}\text{Ca}$ isotopes were successfully measured for the $4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D2) ionic transition and the extracted isotope shifts relative to the ^{40}Ca show an excellent agreement with the literature values. This system is now ready to be applied at the radioactive ion beam facility, such as BRIF, and has paved the way for further development of the higher-sensitivity collinear resonant ionization spectroscopy.

Keywords: Nuclear properties, Collinear laser spectroscopy, Laser-ablation ion source, Photon detection, Isotope shift

I. INTRODUCTION

Understanding the nuclear structural evolution of short-lived exotic nuclei towards the proton and neutron driplines is one of the main topics in the current nuclear physics research, triggering continuous developments of experimental techniques and theoretical approaches [1, 2]. The static properties of the ground and isomeric states of unstable nuclei are of indispensable importance for the study of exotic nuclear structure [3–5], and also represent a stringent test of the different nuclear models [6, 7]. Collinear laser spectroscopy (CLS) has been proven to be one of the powerful tools to access multiple nuclear properties of the ground and isomeric states of exotic nuclei [8, 9]. This is realized by probing the hyperfine structure (hfs) and isotope shift resulted from the interaction between the atomic nucleus and surrounding electrons, which allow to precisely extract the nuclear spins (I), magnetic dipole moments (μ), electric quadrupole moments (Q_s), and changes in the mean square charge radii ($\delta\langle r^2 \rangle$) of an isotopic chain in a nuclear model-independent way.

Along with the observation of unexpected nuclear phenomena in short-lived exotic nuclei, the upgrade of existing and the development of next-generation radioactive ion beam (RIB) facilities are motivated with the aim to produce more exotic radioactive beams. Meanwhile, considerable efforts have been made to continuously enhance the experimental sensitivity and precision of the CLS method [10, 11], facilitating the unceasing studies of exotic nuclei. Up to now, this experimental technique has been established at different

RIB facilities, e.g. ISOLDE/CERN [12], IGISOL/JYFL [13], ISAC/TRIUMF [14], NSCL/MSU [15] and ALTO [16], yielding major inputs for nuclear structure study and providing important benchmark for the development of state-of-the-art nuclear theory [7, 17–19].

Two operational RIB facilities are available in China, i.e. HIRFL (PF-type) at IMP of Lanzhou [20] and BRIF (ISOL-type) at CIAE of Beijing [21], which have played a significant role in the nuclear physics research [20–22]. To gain access to more rare isotopes, two next generation RIB facilities, high intensity heavy ion accelerator facility (HIAF) [23] and Beijing isotope-separation-on-line neutron-rich beam facility (BISOL) [24] are under construction and planned, respectively, offering new opportunities for nuclear physics studies in the near future. However, the well-established CLS technique has so far not been implemented at these domestic RIB facilities. Therefore, in order to take the full advantages of the short-lived isotopes available at these RIB facilities, we have, as a first stage, developed a CLS device combined with an offline laser ablation ion source in order to fully master the laser spectroscopy technique for nuclear properties measurement. This integrated system also paves the way for the further development of high-sensitivity collinear resonance ionization spectroscopy, which allows for the exploration of the more exotic cases.

Here, we present the details of the newly developed CLS apparatus, including the laser ablation ion source with up to 30 keV HV platform, the beamline with the photo detection system, the laser system, and data acquisition system. Thanks to the ion bunches (with about 10 μs temporal length) offered by the laser ablation ion source, the first commissioning experiment was successfully performed to probe the $4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D2) ionic transition of calcium. High-resolution optical spectra of four stable $^{40,42,44,48}\text{Ca}$ isotopes were measured, reaching a typical linewidth of ~ 55 MHz, being comparable with the well-established standard CLS setup worldwide [6, 15, 25]. Isotope shifts ($\delta\nu^{40,A}$) of $^{42,44,48}\text{Ca}$ isotopes relative to ^{40}Ca reference isotope were extracted, which are

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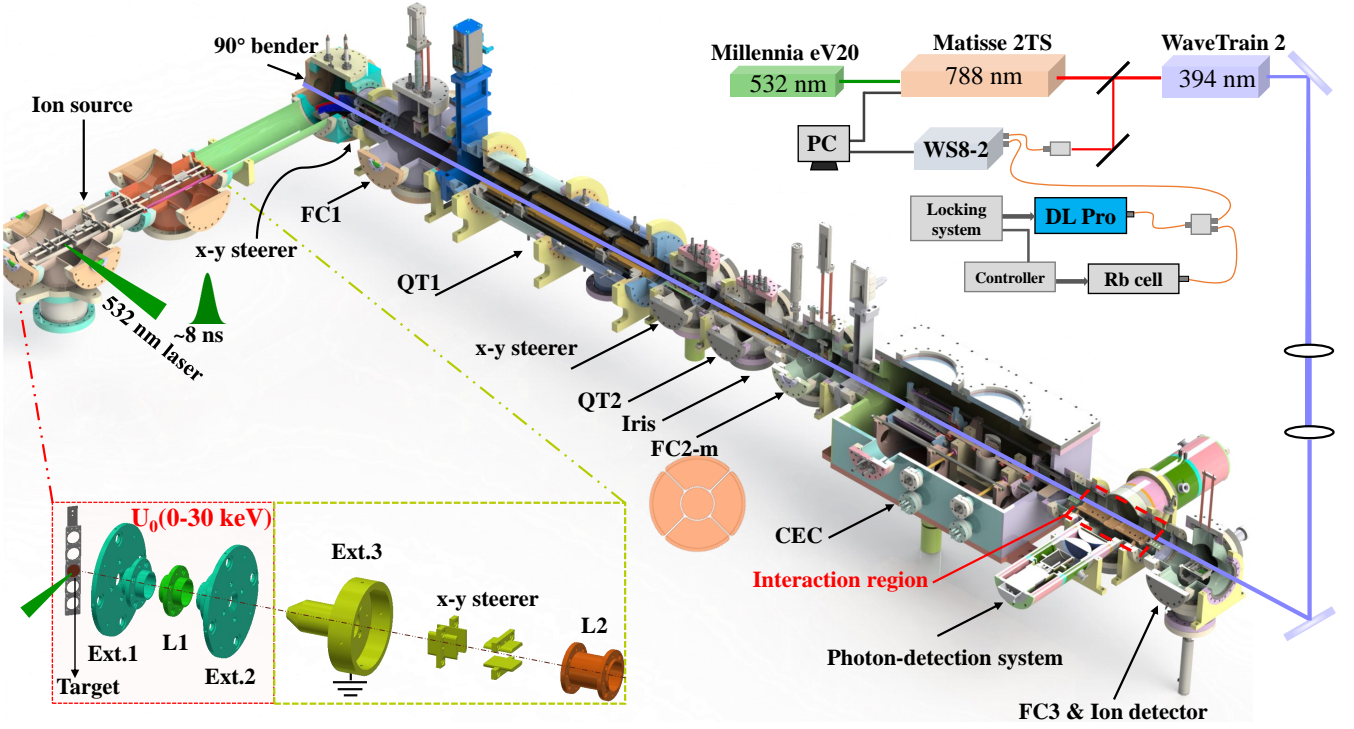


Fig. 1. Schematic view of the CLS device, the electrodes of laser ablation ion source (bottom left inset) and the adopted laser system (top right inset). The stable ion bunches with up to 30 keV energy can be delivered into the beamline, and overlapped with a continuous-wave frequency tunable laser beam in an anti-collinear geometry. The ion beam velocity or laser frequency is tuned to resonantly excite the ions in the interaction zone. The fluorescence photons emitted from the excited ions are collected by the photon detection system. See text for more details.

in excellent agreement with the literature values [6, 26, 27].

will be introduced in the following sections.

II. COLLINEAR LASER SPECTROSCOPY SYSTEM

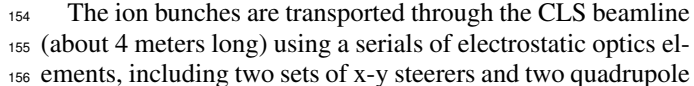
A. Laser ablation ion source

Figure 1 presents a detailed sketch of the CLS apparatus, laser ablation ion source and the laser system. The whole setup is constructed according to the ConFlat (CF) standard for a high vacuum condition, which is currently $\sim 10^{-8}$ mbar, permitting a subsequent upgrade of high-sensitivity collinear resonance ionization spectroscopy setup. With the current system, the hfs spectra of the stable ion beams produced by the ion source can be measured with high-resolution. In brief, the bunched ion beam extracted from the laser ablation target (solid material) is accelerated up to 30 keV. After a 90° electrostatic deflection, the ion bunch is delivered into the CLS beamline, where it is anti-collinearly overlapped with a continuous wave (cw) laser beam. The velocity of the ions can be tuned by applying a scanning voltage to the electrode in the interaction region. The fluorescence photons emitted from the laser excited ions are collected and recorded by the photon detection system and data acquisition system, as a function of the tuning voltage. In the case of probing a transition from neutral atoms, a charge exchange cell (CEC, see Fig. 1) will be required, and the scanning voltage will be applied to an electrode upstream of the CEC. This part will however not be introduced here as we will focus mainly on the Ca ion beam measurement. The functional details of the individual parts

The laser ablation ion source is developed to produce stable ion beams of a wide range of elements with an energy of up to 30 keV [25, 28]. By using a pulsed-laser ablation process, bunched ion beams with a typical temporal length of about $10 \mu\text{s}$, a high ion intensity and a low energy spread can be generated. These features of the ion beam are suitable for the optimization and commissioning of the system of CLS and future resonance ionization spectroscopy, for the development laser excitation and ionization schemes [28], and for the measurement of atomic hfs parameters of stable isotopes [29].

The inner structure of the ion source is shown schematically in Fig. 1 (left bottom inset). A 532-nm Nd: YAG laser (Litron TRLi 250-100), operated at 100 Hz repetition rate and with a pulse width of ~ 8 ns, is employed for the ablation of the solid target. The target holder, floated at the acceleration (platform) potential (U_0 : up to 30 keV) is tilted at 45° with respect to the laser beam and ion beam. The pulsed laser beam is focused onto the target material with a diameter of approximately 1 mm. The ions generated at the ablation area are extracted and refocused with a multiple-step extraction system, namely the first extraction electrode (Ext.1), the first einzel lens (L1) and the second extraction electrode (Ext.2).

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The ion bunches are transported through the CLS beamline (about 4 meters long) using a series of electrostatic optics elements, including two sets of x-y steerers and two quadrupole

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As shown in Fig. 3, each detection unit is composed of two aspheric lenses (namely a telescope), and a PMT. These lenses, with a diameter of 100 mm (N-BK7), have a transmittivity of $\sim 90\%$ over a wide range of wavelength 350-1000 nm, which are used to guide the laser-induced fluorescence photons onto the sensitive area of the PMT. A PMT (R943-02, Hamamatsu) assembled with a socket (E2762-506

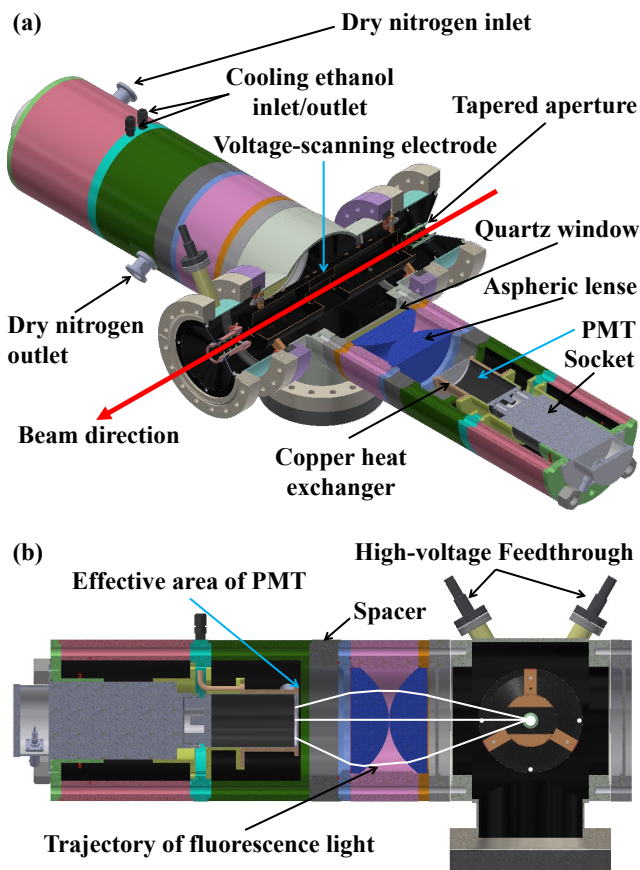


Fig. 3. (a) Photon detection system consisting of two sets of detection units, each including a quartz window, two aspheric lens and a photon-multiplier tube [31]. (b) A schematic view of one detection unit.

Hamamatsu) is employed to record the light. This PMT is featured with the ultra low dark count rate (typical 20 s^{-1} at -20°C) and the wide spectral response (160-930 nm), but with a moderate sensitive area of $10 \times 10 \text{ mm}^2$. The quantum efficiency of this PMT at 394 nm is $\sim 20\%$. Using two detection units, a total geometrical efficiency for fluorescence photon is simulated to be $\sim 15\%$ [32]. The distance between the lens and the PMT is adjustable, which is realized by changing the thickness of the spacer (e.g. 27.3 mm for a wavelength of 394 nm), in order to ensure an optimal geometrical efficiency for various wavelength associated with specific ionic/atomic transitions.

To reach a low dark count rate (dozens of counts per second), the R943-02 PMT needs to be maintained at the temperature of $-20 - -30^\circ\text{C}$. To cool the GaAs (Cs) photocathode, the head of the PMT is covered by a spiral copper tube, named as copper heat exchanger in Fig. 3, through which the ethanol is circulated by a refrigerant circulator (ECO RE 630S, LAUDA). In order to prevent the formation of the frost/ice in the glass window of the PMTs resulted from the low-temperature environment, the dry nitrogen gas is circulated in the system for few minutes before cooling. The above-mentioned spacer, made of PVC, is also used as

a thermal isolation between the PMT and lens, which can effectively avoid the damage of the lens due to the expansion and contraction caused by temperature variation. Based on all these treatments, the achieved typical background rate is about 1 kHz for a 1.2-mW laser beam (about 6-mm diameter). It is worth mentioning that the whole detection unit (include the PMT) is proved to be robust and stable after frequent assembly and disassembly during the commissioning.

D. Laser system

The laser system used for this CLS setup is partly shown in the top right inset of Fig. 1. The continuous-wave (cw) titanium-sapphire (Ti:Sa) laser (Matisse 2TS, Sirah Lasertechnik) is pumped by a 20 W 532 nm laser (Millennia, Spectra-Physics), which can also be converted into the Dye system via an Exchange Kit (TIDYECW, Sirah Lasertechnik). This system provides a laser beam with a wavelength range of 650-1020 nm, which can be further frequency-doubled with the Wavetrain 2 (Sirah Lasertechnik), generating the 2nd harmonic light covering the wavelength range of 325-500 nm. The Matisse cavity can be actively stabilized by the equipped reference cell, reaching a narrow linewidth of $< 50 \text{ kHz}$. A high-precision wavelength meter (HighFinesse WS8-2) is used to measure the real-time frequency of the fundamental cw light.

The wavelength meter can also be used to perform a long-term stabilization of Matisse cavity through a built-in digital interface. To address the drift problem of the wavelength meter (about several MHz per day), caused by temperature and pressure fluctuations, a commercial saturation absorption spectroscopy unit is introduced. This unit consists of a tunable Diode laser (DLPRO780, TOPTICA Photonics AG), a temperature controlled vapor cell filled with K, Rb, and Cs (COSY), as well as the control and locking electronics. The frequency of diode laser is locked to one of the hyperfine components of the available alkali atom (e.g. ^{87}Rb). The frequency-locked diode laser is then used to calibrate the wavelength meter when it functions as a long-term stabilization of the Matisse [33], or to correct the drift of the wavelength meter by recording both frequency-scanned Matisse laser and frequency-locked Diode laser [34].

E. Electronics and data acquisition system

Two signals from the PMTs are amplified by a fast-timing amplifier (ORTEC FTA820A) and then discriminated by a constant fraction discriminator (CFD, CAEN model N605). After converting to TTL logic signals, they are sent to different channels of a ChronoLogic TimeTagger4-2G time-to-digital converter (TDC) with a 500 ps time resolution. Each event labeled by a timestamp could be tracked to obtain the time of flight (TOF) spectrum of the ion bunch. A master TTL signal with 100 Hz repetition rate (a period of 10 ms), labeled as $T_0 = 0 \text{ } \mu\text{s}$ and generated by a Quantum Composers 9528 (QC9528) digital-delay pulse generator, is used to externally

trigger the 532-nm Nd: YAG laser. The laser pulse used for the ablation ion sources arrives $490 \mu\text{s}$ later than T_0 . Therefore, after taking into account of the flight time of the ion bunch from the ion source to the detection region, the start time (trigger) and the time window for the TDC are set to be $T_1 = T_0 + 498 \mu\text{s}$ and $\Delta T = 100 \mu\text{s}$, respectively. This time window covers the time period when the bunched beam is traversing the photon detection region. A narrow time gate, $\sim 10 \mu\text{s}$ corresponding to the width of the ion bunch, can be further applied to reduce the background counts during the offline analysis (more discussion in Sec. III).

The optical hfs spectrum of an isotope is measured by applying a scanned voltage (ΔU) to the electrode tube in the interaction detection region, while the laser frequency is fixed and stabilized with the wavelength meter. A scanned voltage between -1 keV and $+1 \text{ keV}$ can be provided by a DC amplifier (Kepco Model BOP 1000DM) with a gain of 100 and a long-term stability of $< 0.01\%$ over 8 hours, which is controlled by a USB device (USB-3106, Measurement Computing). A real-time measurement of the ΔU applied to the electrode tube and the starting potential U_0 of the ion beam are realized by a Keysight 34470A digital multimeter.

A program written in Python is used for the acquisition system. This program integrates the functions of logging the photon events from the TDC, controlling the scanning voltage via the USB device, recording the frequencies of Matisse and Diode laser via the wavelength meter, reading the scanned voltage and starting potential of the ions through the multimeter, as well as the display of the results via the graphical interface.

III. COMMISSIONING TEST AND RESULTS

To validate the performance of the CLS system, we performed the first commissioning experiment on natural $^{40,42,44,48}\text{Ca}$ isotopes by probing the $4s^2S_{1/2} \rightarrow 4p^2P_{3/2}$ (D2) ionic transition. The stable beams were produced by ablating a calcium target using the 532 nm laser (about 1 mm beam diameter on the target) with an approximately 1.5 mJ/pulse power. In this test experiment, the extracted ion bunches were accelerated to 20 keV and delivered to the CLS beamline. The ion beam was anti-collinearly overlapped with the frequency-fixed cw laser beam (Fig. 1). The laser frequency was stabilized by the wavelength meter, which was calibrated by the diode laser locked to one hyperfine component of the ^{87}Rb atom. The used laser power is about 1.2 mW, and the diameter of the laser spot is $\sim 6 \text{ mm}$. The velocity of Ca ion was tuned in the interaction region by applying a scanning voltage (ΔU) to the electrode tube (Fig. 3). As a result, in the anti-collinear configuration, the Doppler-shifted laser frequency ν experienced by the calcium ion beam can be calculated in the rest frame as:

$$\nu = \nu_0 \times \frac{\sqrt{1 - \beta^2}}{1 - \beta} \quad (1)$$

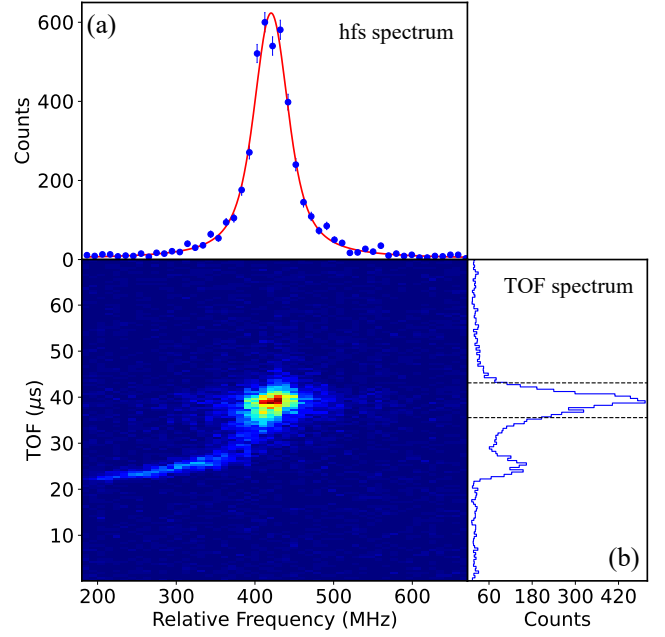


Fig. 4. Color-coded two-dimensional spectrum, TOF vs frequency, for $^{40}\text{Ca}^+$. The projection of this plot onto x-axis (relative laser frequency) and y-axis (time of flight: TOF) leads to the hfs spectrum (a) and TOF spectrum (b). A typical temporal gate of $\sim 10 \mu\text{s}$, as indicated by the dotted lines in (b), is applied to obtain the hfs spectrum as displayed in (a). The hfs spectrum is fitted with a Voigt line profile (red line).

with

$$\beta = \sqrt{1 - \frac{m^2 c^4}{(eU + mc^2)^2}} \quad (2)$$

where ν_0 is the fixed laser frequency, U is the total potential ($U = U_0 + \Delta U$) and m is the mass of the Ca ion. By counting the emitted fluorescence photons from the resonantly excited ions as a function of the tuning voltage (ΔU), the optical spectra for Ca isotopes were obtained.

Figure 4 presents a typical two-dimensional spectrum for relative laser frequency and TOF. The projection of the photon counts onto the x-axis and y-axis lead to the hfs spectrum (Fig. 4(a)) and TOF spectrum (Fig. 4(b)), respectively. The typical temporal length of the ion bunch is about $10 \mu\text{s}$, but with a visible tail in the higher energy side (the shorter TOF side), as shown in the TOF spectrum. Such higher energy tail is related to the field distribution within the plasma plume of the ablation process, as described in Refs. [25, 28]. A TOF correction method [28, 29] can be applied to compensate the higher energy component of the ion bunches, which leads to the similar result as that achieved by simply gating on the main TOF peak, as indicated by the dotted lines in TOF spectrum. This low probability tail is nearly invisible when a weak ion beam is adopted, e.g. the ion beam current of $< 1 \text{ pA}$ as that for $^{42,44,48}\text{Ca}$.

The high-resolution hfs spectra of $^{40,42,44,48}\text{Ca}$, obtained by gating the TOF window as indicated in Fig. 4(b), are shown in the insets of Fig. 5, which are fitted using a Voigt

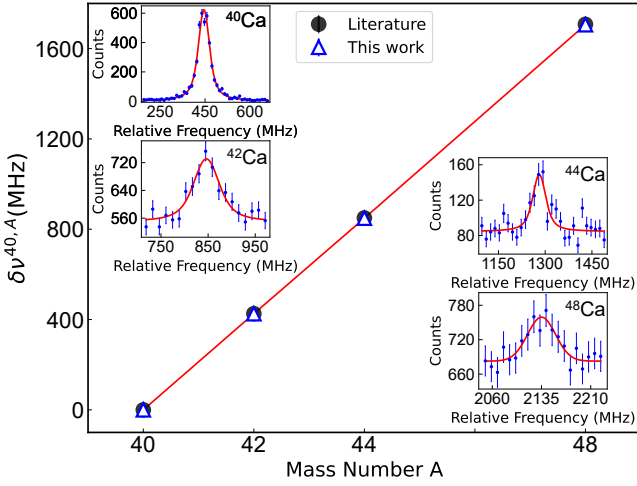


Fig. 5. Presently measured isotope shifts of $^{40,42,44,48}\text{Ca}$ for the $4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D2) ionic transition in comparison to literature values [26]. The optical spectra for $^{40,42,44,48}\text{Ca}$ shown in the insets are fitted with Voigt line profiles (red lines).

TABLE 1. Isotope shifts (in MHz) of $^{42,44,48}\text{Ca}$ isotopes relative to ^{40}Ca measured for the $4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D2) ionic transition. These results are compared with the literature values [26, 27]. Note that the isotope shifts from Ref. [27] are from a dedicated experimental setup for high-precision measurement.

	This work	Ref. [26]	Ref. [27]
40	-	-	-
42	426.9(29)	426.4(15)(10)	426.04(15)
44	848.8(37)	850.1(10)(19)	850.09(14)
48	1705.7(39)	1710.6(35)(39)	1707.58(16)

profile (a convolution of Gaussian and Lorentzian). The full width at half maximum (FWHM) of the spectra from the Voigt fit is about 55 MHz, which is comparable to the same type of CLS measurements worldwide [6, 15, 25]. Considering the natural line width of about 25 MHz (main contribution for the Lorentzian component Γ_L) for the $4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D2) ionic transition, the maximal Gaussian contribution (Γ_G) of the FWHM is about 40 MHz, which is mainly attributed to the energy spread of the ion beam. Assuming an energy spread δE for the total potential U of ion beam, the

resulted Doppler broadening of the spectral line will be:

$$\delta\nu = \nu_0 \times \frac{\delta E}{\sqrt{2eUm}c^2}. \quad (3)$$

Thus, the ~ 40 MHz Gaussian (Γ_G) contribution in linewidth corresponds to an energy spread of ~ 2 eV for the ion beam, which is mainly attributed to the fluctuation of the Heinzinger power supply (~ 200 mV: 4 MHz), Kepco DC amplifier (~ 20 mV) and the field distribution at the position of the ablation target. From these high-resolution optical spectra (shown in the insets of Fig. 5), isotope shifts of $^{42,44,48}\text{Ca}^+$ relative to reference isotope $^{40}\text{Ca}^+$ are extracted, which are in good agreement with the literature values [26, 27], as displayed in Fig. 5 and summarized in Table 1.

IV. SUMMARY AND PROSPECTS

In summary, a collinear laser spectroscopy apparatus integrated with a laser ablation ion source and a frequency tunable laser system, have been implemented at Peking University, aiming at studying nuclear properties of unstable nuclei at the domestic RIB facilities. The ion source was designed to provide the bunched stable ion beam with a beam energy up to 30 keV, which was commissioned with the 20 keV bunched stable calcium ion beam. The typical temporal width of the ion bunch was determined to be about 10 μs . Combined with an anti-collinear laser of 394 nm, high resolution hfs spectra were measured for stable $^{40,42,44,48}\text{Ca}$ isotopes, reaching a narrow linewidth of about 55 MHz (FWHM), which is comparable to the same type of CLS setups worldwide. The Gaussian component of the linewidth (FWHM) was determined to be about 40 MHz, corresponding to an energy spread of ~ 2 eV for the stable calcium ion beam. Isotope shifts ($\delta\nu^{40,A}$) of $^{42,44,48}\text{Ca}$ relative to the reference $^{40}\text{Ca}^+$, extracted from the obtained hfs spectra, are in excellent agreement with the literature, demonstrating the overall satisfactory performance of this CLS system.

Based on the successful implementation and operation, the entire system is now ready to be applied to study unstable isotopes at the RIB facilities, such as BRIF of CIAE. This kind of online experiment is scheduled and will be performed in a short term. In addition, further exploration and developments of the CLS technique are also planned in the coming steps, e.g. the application of system for atomic hfs spectrum measurement using a charge exchange process and the development of the system towards a resonant ionization spectroscopy measurement. A mass separator and a radio-frequency quadrupole cooler/buncher can also be incorporated into the system, in order to provide ion beam bunch with a better time structure.

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